

On search for nuclear Schiff moment in liquid xenon

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Abstract

A parameter of the P,T-odd Hamiltonian characterizing interaction of the nuclear Schiff moment with the gradient of electronic density on the Xe nucleus is calculated for an isolated Xe atom and for liquid xenon. We use more realistic model of liquid medium than the spherical cell model used in [B.Ravaine and A.Derevianko Phys. Rev. A **69**, 050101(R)(2004)]. Qualitatively different results for enhancement of the P,T-odd effect in liquid xenon are obtained when polarization of the medium is taken into account. Thus, proper choice of the liquid phase model is crucially important even for calculation of the properties dependent mostly on the electronic density near a nucleus.

INTRODUCTION.

Schiff moment is a space parity (P) and time invariance (T) violating (P,T-odd) nuclear moment that is expected to exist because of P,T-odd interactions between nucleons and/or existence of uncompensated contribution from the permanent electric dipole moment (EDM) of the unpaired nucleon, enhanced (or, in principle, suppressed) by nucleus core polarization [1]. The conventional choice for experiments on search for the Schiff moment up to the present time has been heavy atoms or polar diatomic molecules containing a heavy atom because of great enhancement of the P,T-odd effects in such systems. Recently some new approaches to search for the Schiff moment were suggested. Their principal feature is using more complex systems, particularly, solid state [2] and liquid xenon (LXe) [3, 4] have been considered for the corresponding experiments. Though solids and liquids have a number of advantages first of all because of the higher statistical factor in comparison with that for the atomic/molecular beam/cell experiments, reliable calculations of their electronic structure (always required in the experiments for the interpretation of the data in terms of constants of the P,T-violating weak interaction) are, certainly, substantially more complicated than for the diatomic molecules. Even in a closed-shell Van der Waals system such as liquid xenon the choice of the appropriate model for the medium is not trivial for the purpose of reliable theoretical study. For example, in [5] the cell model of liquid xenon was used. In that model the single Xe atom is confined in the spherically symmetric cavity and a density-dependence factor is introduced through variation of the cavity radius. Using the cell model could be justified by the fact that LXe is non-polar liquid with the dielectric constant close to unity. In [5] authors found that the effect of P,T-violation is suppressed in their model for liquid xenon on about 40% in comparison with an isolated (free) atom. However, the influence of polarization of the medium on the P,T-odd property was not studied. The purpose of the current work is calculation of the P,T-odd effect caused by the Schiff moment of the ^{129}Xe nucleus using more realistic models of LXe than the cell model and studying the contribution from polarization of the medium on the P,T-odd property in liquid xenon.

METHODS AND APPROACHES.

Electronic structure calculations of the isolated Xe atom and liquid medium are carried out using two-step method (see [6] and references therein), the most principal features of which are:

1. A two-component electronic (pseudo)wave function is first obtained in calculation with the generalized relativistic effective core potential (GRECP) for Xe with either 8 or 26 explicitly treated electrons, providing proper electronic density in the valence and outer core regions.
2. The proper shape of the four-component molecular spinors in the inner core region of the Xe atom is restored using the non-variational one-center restoration (NOCR) scheme [7, 8, 9] followed by calculations of P,T-odd parameters essentially depending on electronic density near the heavy nucleus.

In the present work, the finite field method [10] is used for the required property calculations in the framework of the relativistic coupled cluster method with single and double cluster amplitudes (RCC-SD) [11, 12]. The latter was chosen because it is known to work well for the closed shell systems.

Generalized correlation-consistent (GC) spin-orbital atomic basis sets [13, 14] are used in xenon electronic structure calculations. GC basis sets were specially optimized for calculation of the “core-type” properties and, besides, were augmented with the polarization d -function optimized with respect to the value of atomic polarizability α_p . As a result, basis set [4s,6p,4d,2f] was constructed for calculations with 8-electron GRECP and [5s,6p,4d,2f] for 26-electron GRECP. All molecular spinors are then restored as one-center expansions on the ^{129}Xe nucleus. The nucleus is modelled as a uniform charge distribution within a sphere with the radius $r_{\text{nucl}} = 6.2 \text{ fm} \equiv 1.17 \times 10^{-4} \text{ a.u.}$ The gaussian expansion of basis sets and results of atomic calculations can be found elsewhere (see <http://qchem.pnpi.spb.ru/Basis/Xe>). To calculate the atomic polarizability and parameter of the P,T-odd Hamiltonian, X (see Eq. (1)), we have applied electric field created by four point electric charges located on z axis. In order to reach sufficient homogeneity of electric field at the xenon cluster the charges $\{-80, 5, -5, 80\}$ a.u. were located in points $\{-100, -50, 50, 100\}$ (a.u., on z axis), thus electric field 0.012 a.u. was created at the coordinate source. Below we will use the atomic units

unless other is stated explicitly. Such electric field is weak enough to neglect higher-order terms in calculations of α_p and parameter X , but sufficiently strong to avoid problems with numerical accuracy. The field homogeneity and strength were checked in the atomic SCF and RCC-SD calculations of α_p with homogenous field and X taking the field 0.0012 (ten times weaker than the originally used one).

The parameter X of the P,T-odd Hamiltonian [9] was calculated for the Xe atom in the presence of electric field:

$$X = \frac{2\pi}{3} \left[\frac{\partial}{\partial z} \rho_\psi(\vec{r}) \right]_{x,y,z=0}, \quad (1)$$

where $\rho_\psi(\vec{r})$ is the electronic density calculated from the wave function ψ . Such form of X is derived from contact form of the P,T-odd interaction of the Schiff moment with the electronic density. In [1] more sophisticated form of P,T-odd interaction was suggested, where the “finite-volume” Schiff moment potential is used. In the atomic Xe calculations it was found that the “finite-volume” form of Schiff interaction does not lead to noticeable changes in the results of the P,T-odd properties calculation in comparison with the contact one [15]. Connection of the parameter X with the parameters typically evaluated in atomic calculations of the EDM induced by the Schiff moment can be derived following [16]:

$$X = \frac{1}{6} \frac{\mathbf{S} \cdot \mathbf{n}}{\mathbf{D}_e \cdot \mathbf{E}},$$

where \mathbf{S} is the Schiff moment of ^{129}Xe nucleus, \mathbf{n} is a unit vector along the molecular axis (which in our case coincides with the direction of external electric field), \mathbf{D}_e is the atomic EDM induced by the nucleus Schiff moment, \mathbf{E} is an external electric field applied to the Xe atom.

Atomic calculation. To analyze the different correlation contribution we have performed calculations of X and α_p for an isolated Xe atom with the different numbers of correlated electrons and the level of correlation treatment. The results are summarized in Table I. Spin-averaged GRECP (AGREP) was used in SCF calculations, while the spin-orbit GRECP component was also included in RCC-S and RCC-SD calculations. The difference between 26-electron GRECP/RCC-S (which is roughly analogous to SCF calculation accounting for the spin-orbit (SO) interaction in the closed-shell case) and all-electron DHF results is within 10%, which can be connected with the incompleteness of basis sets and polarization contributions from inner shells. As the main purpose of the current work is not calculating the P,T-odd effects with very high precision but rather investigating the influence of the

TABLE I: Calculated parameter X and scalar polarizability α_p for the ^{129}Xe single atom, compared with values from [16]. Relativistic coupled cluster methods with single (RCC-S) and single and double cluster amplitudes (RCC-SD) are used with 8- and 26-electron Xe GRECPs. All values are in a.u.

	8 electrons		26 electrons			All-electron ^a		
	AGREP/SCF	GRECP/RCC-SD	AGREP/SCF	GRECP/RCC-S	GRECP/RCC-SD	DHF	TDHF	Experimental
X	238	213	186	176	173	165	213	
α_p	26.7		26.6	26.6	27.2	26.9	27.0	27.16 ^b 27.815 ^c

^a Four component atomic calculations by Dirac-Hartree-Fock method (DHF) and time-dependent DHF (TDHF).

^b Reference [17].

^c Reference [18].

liquid medium on the P,T-odd effect(s) we use 8-electron GRECP in subsequent calculations to reduce computational expenses. It is seen from Table I that contribution to the X value from the SO effects is only about 5%, thus we are not calculating further RCC-S values separately and rather going from AGREP/SCF right to GRECP/RCC-SD values accounting for both correlation and SO-effects. Note, however, that contribution of the SO interaction taken into account at the GRECP/RCC-SD or GRECP/RCC-S calculation (for the “outer parts” of explicitly treated shells) is different from the SO contributions taken into account at the AGREP/SCF stage (for the inner shells excluded from the GRECP calculations since they were treated as SO-split atomic spinors at the GRECP generation) and at the NOCR stage (for the “inner parts” of the shells explicitly treated within GRECP calculations).

Liquid xenon model. In liquid xenon calculations we used the simple cubic crystalline lattice (see Fig. 1). It is known (e.g., see [19]), that the decrease of the density in liquid xenon as compared to the solid state phase is mainly due to decreasing the average number of the neighboring atoms (i.e. introducing vacancies) according to the ratio $\rho_{\text{liq}}/\rho_{\text{sol}}$, where ρ_{liq} (ρ_{sol}) is the density of liquid (solid) xenon. On the other hand, at atmospheric pressure $\rho_{\text{liq}}/\rho_{\text{sol}} \approx 0.83$, thus, the lattice model can be used for the liquid phase description without introducing principal error. Though in solid xenon the crystalline lattice is the face-centered cubic one (FCC), there are some reasons to use simple cubic (SC) lattice model. In such lattice the number of neighboring atoms is reduced twice in comparison with the FCC lattice, nevertheless, the minimal number of the atoms which required for adequate analysis of the

effects of the liquid medium is kept. As it will be seen from the further discussion, the atoms which are in the plane perpendicular to the electric field and going through the central Xe atom practically do not influence the value of X . Thus, on the one hand, high-symmetry SC lattice allows us to analyze the effect of the liquid medium with minimal computational efforts — the only six neighbours reproduce the volume effect reasonably well whereas the number of explicitly treated electrons is reduced almost twice, and, on the other hand, it reflects the most principal features of the liquid medium. Besides, high symmetry of the SC lattice simplifies the analysis of different density- and polarization-dependence effects keeping reasonable computational expenses within the C_{2v} symmetry implemented in the code used by us. Electric field modelled by four point charges described in section “Methods and approaches” was applied to the cluster of seven xenon atoms. We used two geometries of relative arrangement of the Xe atoms and electric field, both geometries are shown on Fig. 1. The other geometries were not considered because they have the symmetry which is not implemented in our codes or require to use lower symmetry that leads to high computational expenses. Blue color means that all explicitly taken eight electrons of the given Xe atom were frozen by the level-shift technique [20] after the atomic spin-free AGREP/SCF calculations, while red color means involving all eight electrons to SCF and RCC procedure. Two series of calculations were performed denoted below as **I** and **II**. In series **I** all electrons of all atoms but the central Xe atom were frozen after the atomic AGREP/SCF calculations (Fig. 1) In series **II** electrons in the 1st geometry were frozen only for atoms which are not on axis z (Fig. 2). The interatomic distance R_l in the lattice cell (lattice constant) was changed from 6.0 a.u. to 20 a.u. The value of $R_l = 6.6$ corresponds to the elementary *lattice* cell of the *same volume* that was taken in [5] for the *spherical* cell. The more realistic value for the equilibrium lattice constant R_l is connected with the parameters of the FCC lattice for the solid xenon density (in this case the distance between the closest neighbors in the FCC lattice is $R_{l-FCC} = 8.6$). As explained above, we, however, use the model of simple cubic lattice with $R_l = n^{-1/3} = 8.0$, where n is the density of liquid xenon (for the solid xenon density the corresponding constant would be $R_l = 7.5$). In any case, as we will see further, the reasonable choice of such or another R_l is not principal for our conclusions.

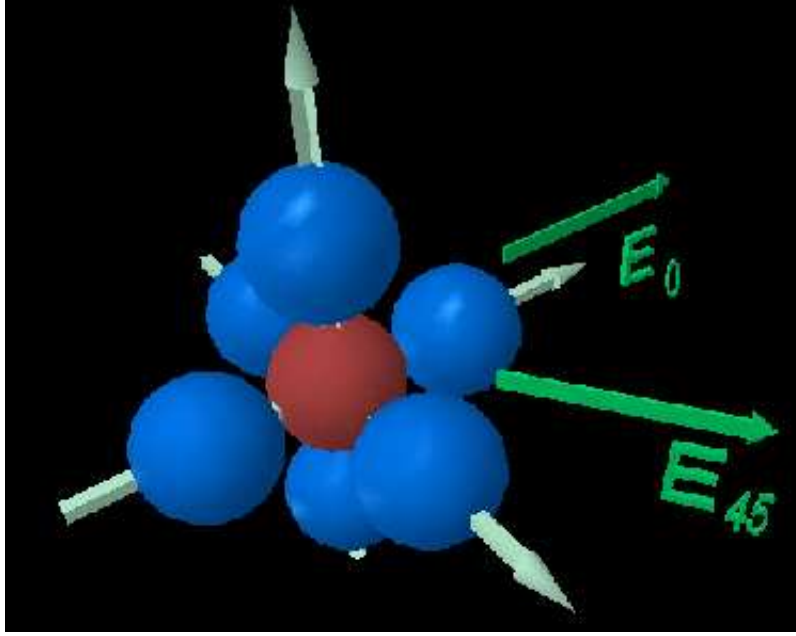


FIG. 1: Series **I** lattice cell geometry and electric field. In the 1st geometry electric field E_0 is directed along z axis. In the 2nd geometry electric field E_{45} is directed under 45° to z axis.

RESULTS AND DISCUSSION

Series I: The results of AGREP/SCF and GRECP/RCC-SD calculations are presented on Fig. 3. Qualitative agreement with the results of [5] is seen from that picture, though the suppression of P,T-odd effect in liquid phase is only about 20%, in contrast to 40% obtained in [5]. An interesting peculiarity which is seen on Fig. 3 is the increase of the X value when $R_l \leq 7.5$. The natural reason for such behavior can be the increase of the amplitude of the wavefunction on the Xe nucleus due to compression of the tail of the electronic density in the valence region. For $R_l > 7.5$ the wavefunction amplitude increasing on the nucleus is less important than effect of suppression of the polarizability of the Xe atom in the “frozen” medium. Actually, the same is observed in the spherical cavity model (where polarizability of the valence shells is suppressed even stronger) that results in decreasing the value of the P,T-odd effect. We have seen very weak dependence (within 1 %) of X value from either 1st or 2nd geometry used. In further calculations we used only the 1st geometry, such choice for series **II** is explained below.

Series II: The results of AGREP/SCF and GRECP/RCC-SD calculations are presented on Fig. 4. The test calculations were performed first with $R_l = 7.5$ to estimate in-

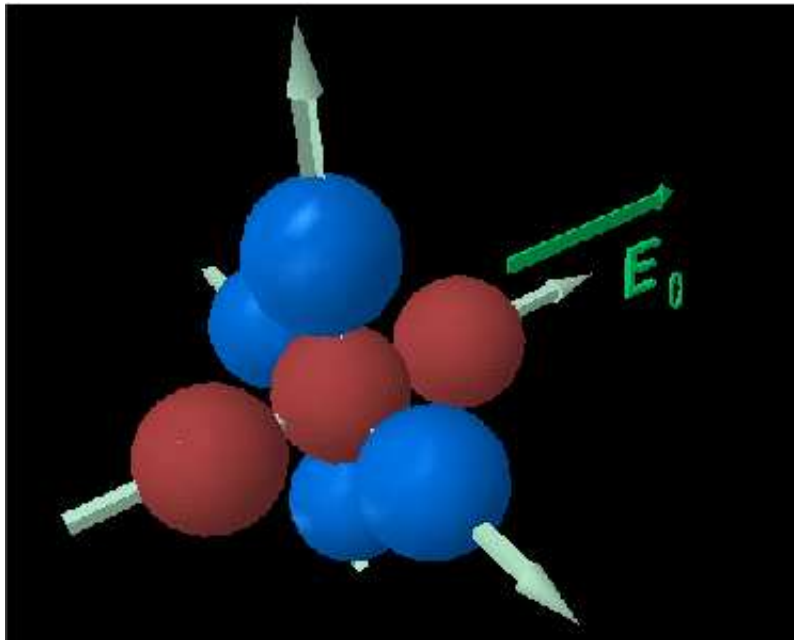


FIG. 2: Series **II** lattice cell geometry and electric field.

fluence of the neighboring atoms. It turned out that accounting for polarization/correlation effects for the atoms which are not on z axis (that is directed along the electric field) practically does not change the value of X , that looks rather natural. Thus, in subsequent calculations we accounted only for polarization/correlation of the electrons belonging to the atoms on z axis. Reduced GC basis sets described in section “Methods and approaches” were used — on the central Xe atom GC basis was $[4s,6p,4d]$ and on two neighbor atoms basis sets were reduced to $[4s,6p]$. Though, formally, $\{s,p\}$ basis set is not good for description of polarization/correlation effects on the atom, functions from the other atom are partially capable to account for those effects.

One can see that accounting for polarization of the neighboring atoms leads to crucial change in the enhancement of the Schiff moment in liquid xenon. The P,T-odd effect is practically *not suppressed* but rather *enhanced* in liquid xenon.

The physical reason for such behavior is interference of the polarization of the liquid medium and the density-dependence effects (compression of the electronic density in the valence region). The following basic interpretations are the most reasonable to consider for the polarization contribution:

1. the tails of electronic density from *neighboring* atoms penetrate to the inner core of the

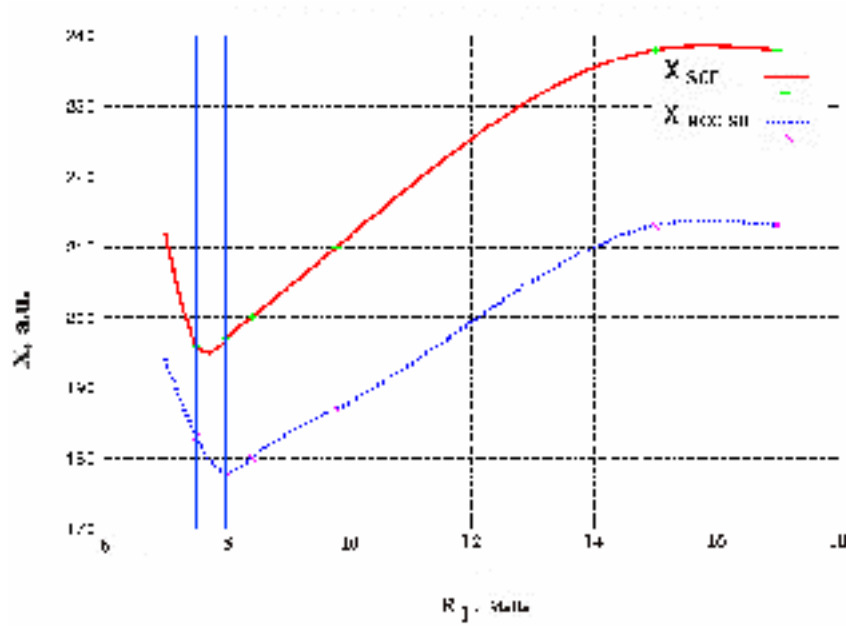


FIG. 3: Series *I* X value as a function of the lattice cell size. X_{SCF} curve corresponds to AGREP/SCF result, $X_{\text{RCC-SD}}$ stands for GRECP/RCC-SD results. Vertical lines are the functions $R_l = 7.5$ and $R_l = 8.0$, see section “Methods and approaches”.

central Xe atom. Due to polarization effects the contributions to the gradient of the electronic density on the central Xe nucleus (which X is proportional to, see Eq. (1)) from neighbors do not compensate each other;

2. *own* electronic shells of the central Xe atom interact with the outermost electronic shells of neighboring atoms, thus affecting the X value. As outer shells of neighboring atoms are polarized in the electric field, their polarization influences the polarization of the outer shells of the central Xe atom. Also, the amplitude of the electronic wavefunction could be increasing at the ^{129}Xe nucleus due to compression of the electron wavefunction tails in the valence region whereas the polarization of the isolated atom and the atom in liquid medium could be comparable. Such an effect of the “volume compression” would manifest itself in $1/R_l^3$ -like dependance of the X value from R_l (with R_l for which outermost shells of the neighbour Xe atoms start to overlap) on

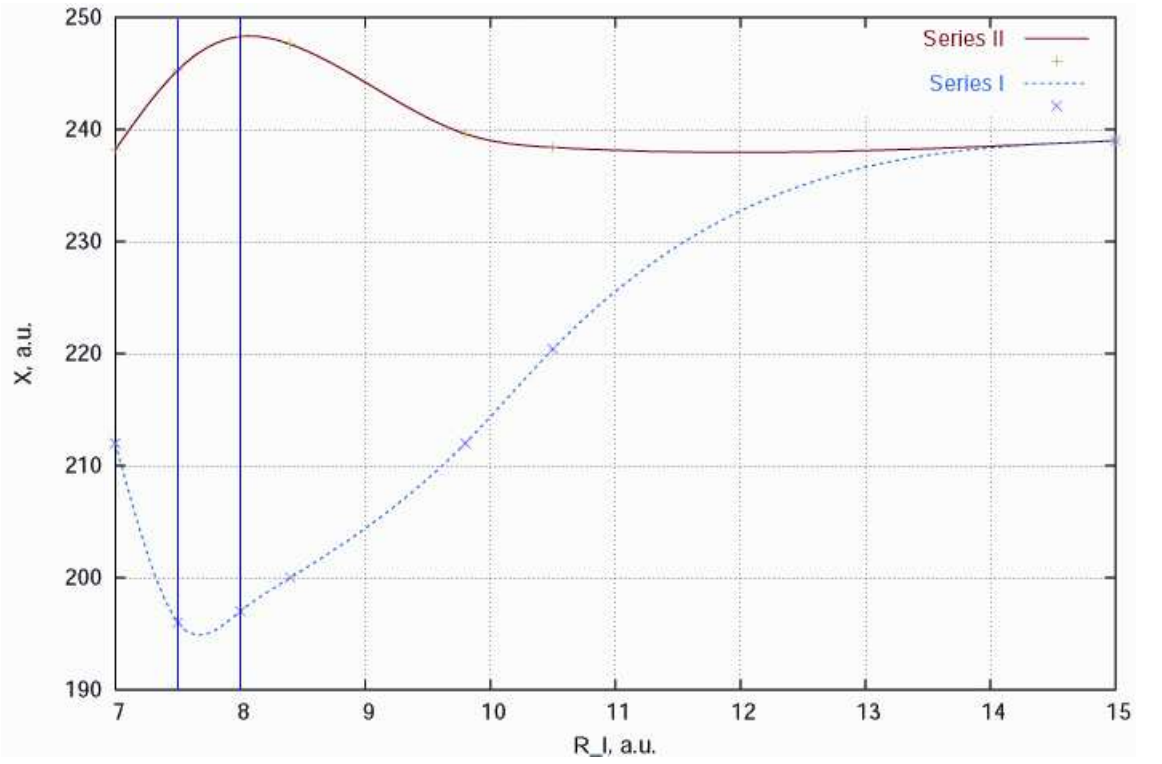


FIG. 4: Series *I* and *II* AGREP/SCF X values as a function of the cell size. Vertical lines are the functions $R_l = 7.5$ and $R_l = 8.0$, see section “Methods and approaches”.

Fig. 4, but, as our analysis show, the dependance $X(R_l)$, ($9.0 < R_l < 10$), is linear. Nevertheless, some combination of the above effects is rather taking place.

Though the above discussed effects are undistinguishable physically (at least for the case of the xenon cluster), from computational point of view they are treated as different ones. The spherical cell model in [5] does not account for the above effects properly.

To clarify the situation we carried out two series of calculations. In the first one we placed three Xe atoms on z axis and the distance between the Xe nuclei was chosen to be 7.5. The electrons of the central atom were frozen, while the electrons of its two neighbors were allowed to relax (polarize) in the external electric field, which strength was 0.012. Thus, the only contribution to X came from the electronic density from the neighboring atoms, as frozen atomic shells of the central atom had definite parity. We obtained $X = 1.4$. On the other hand, the contribution from every neighbor was about $X_s \approx \pm 24$ (sign depends on relative position with respect to the central atom), thus the great compensation of the contributions from the neighboring atoms takes place. In the second series we performed calculation of X value almost in the same geometry as in series **I**. Electrons of neighboring atoms were frozen, while in the central atom eight electrons were correlated. Then the central atom was shifted with respect to the coordinate source on the value Δz , see Fig. 5 (picture **S**).

As one can see from Fig. 5 such geometry simulates polarization of the shells of the neighbor atoms with electric field. The obtained dependance of the X value from ΔZ is given on Fig. 6. To get X as large as in series **II** one has to take $\Delta Z \approx -0.15$. Taking into account atomic Xe polarizability α_p (see Table I) one can estimate that $\Delta Z \approx -0.15$ is compatible with possible value for the shift of the center of charge density of outer p -electrons in the field 0.012. Thus dramatic change of the X value in the lattice model in comparison with the cell model can be rather attributed to the effect of rearrangement of electronic density of the liquid medium.

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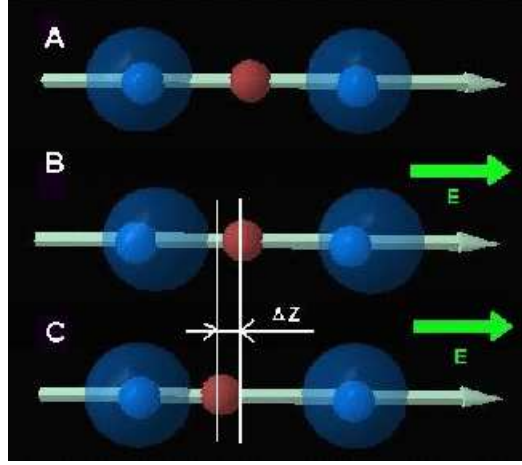


FIG. 5: Simulation of the influence of electric field through lattice geometry. Only atoms on z axis are plotted. Red color designates the nuclei of the central Xe atom (its shells explicitly treated in the calculations are not shown here), outer shells of the neighboring atoms are half-transparent blue, inner shells and nuclei are blue. On picture **A** atoms are without electric field, when center of charge of the outer electronic density coincides with that of the inner shells and nuclei. On picture **B** in the electric field (designated E) the centers of charge of the outer shells are shifted in respect to inner part. On picture **C** outer shells electronic density of the neighboring atoms is shifted so as to superpose their charge center to those of neighboring atoms but keep the relative position to the central Xe atom the same as in case **B**.

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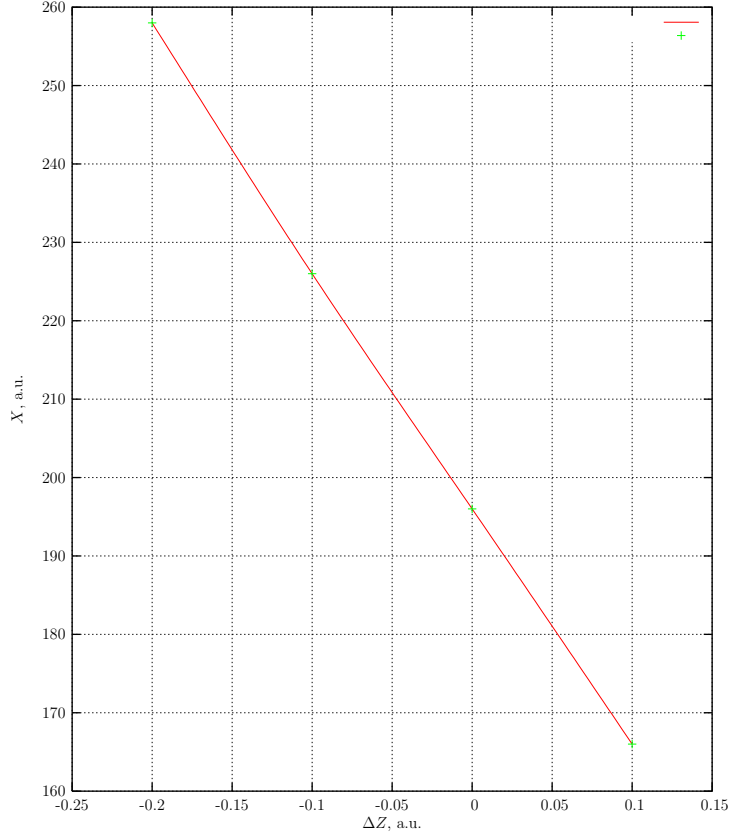


FIG. 6: AGREP/SCF X value as a function of the shift of the central atom.

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